Handout 30

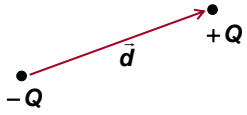
Optical Processes in Solids and the Dielectric Constant

In this lecture you will learn:

- Linear response functions
- Kramers-Kronig relations
- Dielectric constant of solids
- Interband and Intraband contributions to the dielectric constant of solids

Charge Dipole, Dipole Moment, and Polarization Density

A charge dipole consists of a negative and a positive charge separated by some distance:

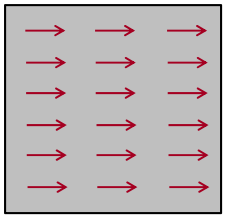


Dipole moment of a charge dipole is a vector $\vec{p}$ such that:

$$\vec{p} = |Q|\,\vec{d}$$

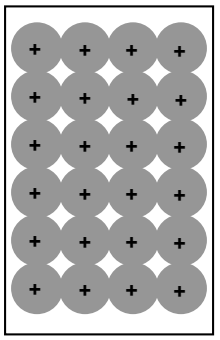
Polarization density vector $\vec{P}$ of a medium consisting of charge dipoles is the product of the number N of dipoles per unit volume (i.e. dipole density) and the strength of each dipole given by $\vec{p}$:

$$\vec{P} = N\vec{p} = N|Q|\vec{d}$$



Dielectric Constant of Non-Polar Materials

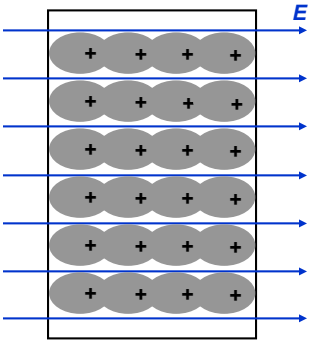
Non-Polar Dielectric Material (Non-polar Insulator or Intrinsic Semiconductor)



\+ +ve nucleus

-ve electron cloud (valence band and core electrons)

Dielectric in an E-field



Material gets **polarized** when placed in an electric field (i.e. develops charge dipoles) because the electron cloud shifts relative to the nuclei

$\vec{P}$ = Polarization density (# of dipoles per unit volume times the strength of one dipole)

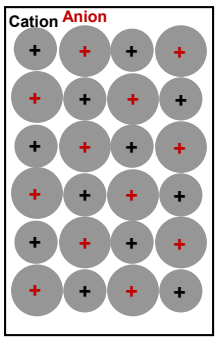
$$\vec{P}_e = \varepsilon_o \chi_e \vec{E}$$

$$\vec{D} = \varepsilon_o \vec{E} + \vec{P}_e = \varepsilon \vec{E}$$

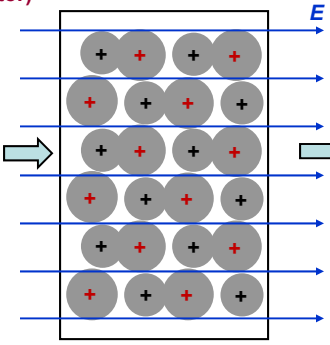
$$\varepsilon = \varepsilon_o (1 + \chi_e)$$

Dielectric Constant of Polar Materials

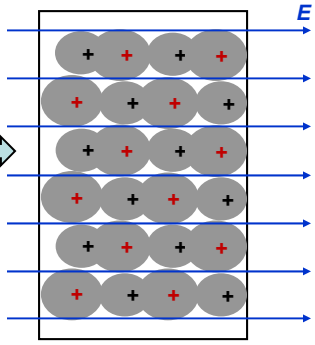
Polar Dielectric Material (Polar Insulator or Intrinsic Semiconductor)



The phonon contribution to Polarization



The total (phonon + electron) contribution to Polarization



In polar materials, material polarization in an E-field has two contributions:

a) The phonon contribution: $\vec{P}_{ph} = \varepsilon_o \chi_{ph} \vec{E}$

b) The electron contribution: $\vec{P}_e = \varepsilon_o \chi_e \vec{E}$

$$\Rightarrow \vec{D} = \varepsilon_o \vec{E} + \vec{P}_e + P_{ph} = \varepsilon_o (1 + \chi_e + \chi_{ph}) \vec{E}$$

$$\Rightarrow \varepsilon = \varepsilon_o (1 + \chi_e + \chi_{ph})$$

Dielectric Constant of Materials: Phonon and Electron Contributions

In general, the susceptibilities are frequency dependent:

$$\varepsilon(\omega) = \varepsilon_o\left(1+\chi_e(\omega)+\chi_{ph}(\omega)\right)$$

Electrons respond much faster than the lattice

If one is working at frequencies that are too small compared to the characteristic frequencies of $\chi_e(\omega)$ then one may make the approximation:

$$\varepsilon(\omega) = \varepsilon_o\left(1+\chi_e(0)+\chi_{ph}(\omega)\right)$$

If we define:

$$\varepsilon(\infty) = \varepsilon_o\left(1+\chi_e(0)\right) \qquad \varepsilon(0) = \varepsilon_o\left(1+\chi_e(0)+\chi_{ph}(0)\right)$$

then for small frequencies:

$$\varepsilon(\omega) = \varepsilon(\infty)+\varepsilon_o\,\chi_{ph}(\omega)$$

Comparing with the expression in handout 19: $\varepsilon(\omega) = \varepsilon(\infty) - \dfrac{nf^2/M_r}{\omega^2-\omega_{TO}^2}$

gives us:

$$\varepsilon_o\,\chi_{ph}(\omega) = -\frac{nf^2/M_r}{\omega^2-\omega_{TO}^2}$$

And we have finally:

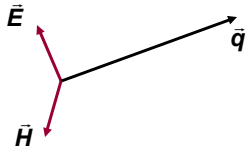
$$\varepsilon(\omega) = \varepsilon_o\left(1+\chi_e(\omega)\right) - \frac{nf^2/M_r}{\omega^2-\omega_{TO}^2} = \varepsilon_o\left(1+\chi_e(\omega)\right) - \omega_{TO}^2\,\frac{\left(\varepsilon(0)-\varepsilon(\infty)\right)}{\omega^2-\omega_{TO}^2}$$

We now find the electronic contribution

High Frequency Dielectric Constant of Solids

Consider a sinusoidal E&M wave of frequency ω propagating in a solid:

$$\vec{E}(\vec{r},t) = \hat{n}\,E_o\cos(\vec{q}\,.\,\vec{r}-\omega\,t) = \text{Re}\left\{\vec{E}(\vec{r},\omega)\,e^{-i\,\omega\,t}\right\}$$



Where the electric field "phasor" is:

$$\vec{E}(\vec{r},\omega) = \hat{n}\,E_o\,e^{i\,\vec{q}\,.\,\vec{r}}$$

Similarly, the magnetic field phasor is:

$$\vec{H}(\vec{r},\omega) = (\hat{q}\times\hat{n})\,H_o\,e^{i\,\vec{q}\,.\,\vec{r}}$$

And the two field are related by the two Maxwell equations:

$$\nabla\times\vec{E}(\vec{r},\omega) = i\omega\,\mu_o\vec{H}(\vec{r},\omega) \qquad \text{Faraday's Law}$$

$$\nabla\times\vec{H}(\vec{r},\omega) = -i\omega\,\varepsilon(\omega)\,\vec{E}(\vec{r},\omega) \qquad \text{Ampere's Law}$$

These two equations together give the dispersion relation of the E&M wave:

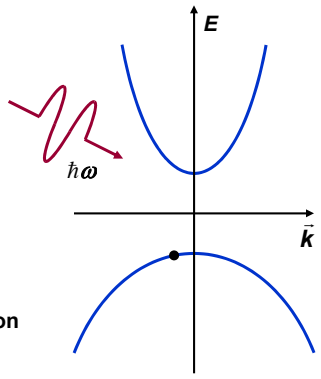
$$\omega = \frac{|\vec{q}|}{\sqrt{\varepsilon(\omega)\,\mu_o}} = q\,\frac{c}{\sqrt{\varepsilon(\omega)/\varepsilon_o}}$$

Strategy to Calculate the Dielectric Constant of Materials

1) Start with the Hamiltonian describing the interaction of the electrons with the electromagnetic field:

$$\hat{H} = \hat{H}_o + \frac{e\,A_o\left[\, e^{i\vec{q}\,.\,\hat{\vec{r}} - i\,\omega\,t} + e^{-i\vec{q}\,.\,\hat{\vec{r}} + i\,\omega\,t} \,\right]}{2m}\,\hat{\vec{P}}\,.\,\hat{n}$$

$$\hat{H}_o \left|\psi_{n,\vec{k}}\right\rangle = E_n\left(\vec{k}\right)\left|\psi_{n,\vec{k}}\right\rangle$$



2) Find out how the electron wavefunctions (i.e. Bloch functions) get modified using standard first order perturbation theory:

$$\left|\psi_{n,\vec{k}}\right\rangle_{\text{new}} = \left|\psi_{n,\vec{k}}\right\rangle + \sum_{m}\sum_{\vec{k}'} c_{m,\vec{k}'}(t)\left|\psi_{m,\vec{k}'}\right\rangle$$

3) From the modified wavefunctions, calculate the electron charge density, and then the dipole density

The above procedure, although doable, is a little complicated and we will use an alternate approach!

Refractive Index of Solids

The refractive index of a material is defined as: $n(\omega) = \sqrt{\frac{\varepsilon(\omega)}{\varepsilon_o}}$

The wave dispersion relation is then: $\omega = q\,\frac{c}{n(\omega)}$

And the electric field phasor can be written as:

$$\vec{E}(\vec{r},\omega) = \hat{n}\,E_o\,e^{i\,\vec{q}\,.\,\vec{r}} = \hat{n}\,E_o\,e^{i\,\frac{\omega\,n(\omega)}{c}\,\hat{q}\,.\,\vec{r}}$$

The refractive index usually has real and imaginary parts:

$$n(\omega) = n'(\omega) + i\,n''(\omega)$$

The electric field phasor is then:

$$\vec{E}(\vec{r},\omega) = \hat{n}\,E_o\,e^{i\,\frac{\omega\,n'(\omega)}{c}\,\hat{q}\,.\,\vec{r}}\,\underbrace{e^{-\frac{\omega\,n''(\omega)}{c}\,\hat{q}\,.\,\vec{r}}}$$

The imaginary part of the index describes wave decay (or wave amplification if gain is present)

Imaginary Part of the Refractive Index and the Loss Coefficient

We have already seen that stimulated absorption results in a wave to decay in a medium (optical loss):

$$\vec{E}(\vec{r},\omega) \propto e^{-\frac{\alpha(\omega)}{2}\hat{q}\,.\,\vec{r}}$$

Where:

$$\alpha(\omega) = \frac{\hbar\omega\left(R_{\uparrow} - R_{\downarrow}\right)}{P}$$

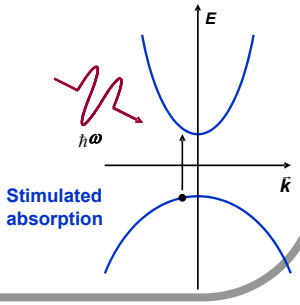
$$= \left(\frac{e}{m}\right)^2 \frac{\pi}{\varepsilon_o n'\omega\, c} \left\langle \left|\vec{P}_{cv}\,.\,\hat{n}\right|^2 \right\rangle 2\times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \delta\left(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega\right)$$

But we also have:

$$\vec{E}(\vec{r},\omega) \propto e^{-\frac{\omega\, n''(\omega)}{c}\hat{q}\,.\,\vec{r}}$$

This means the imaginary part of the refractive index is:

$$n''(\omega) = \frac{c}{\omega}\frac{\alpha(\omega)}{2}$$



High Frequency Dielectric Constant of Solids: Imaginary Part

The refractive index of a material is defined as: $n(\omega) = \sqrt{\frac{\varepsilon(\omega)}{\varepsilon_o}}$

Therefore, using the fact that: $|n''(\omega)| << |n'(\omega)|$

$$\varepsilon(\omega) = \varepsilon_o\, n^2(\omega) = \varepsilon_o\left[\, n'(\omega) + in''(\omega)\,\right]^2 \approx \varepsilon_o\left[\, n'(\omega)\,\right]^2 + i2\,\varepsilon_o n'(\omega) n''(\omega)$$

$$\Rightarrow \varepsilon'(\omega) + i\varepsilon''(\omega) = \varepsilon_o\left[\, n'(\omega)\,\right]^2 + i2\,\varepsilon_o n'(\omega) n''(\omega)$$

This implies:

$$\varepsilon''(\omega) \approx 2\varepsilon_o n'(\omega) n''(\omega) \quad \text{and} \quad \varepsilon'(\omega) \approx \varepsilon_o\left[\, n'(\omega)\,\right]^2$$

Using the expression for the imaginary part of the refractive index we get:

$$\varepsilon''(\omega) = \left(\frac{e}{m}\right)^2 \frac{\pi}{\omega^2} \left\langle \left|\vec{P}_{cv}\,.\,\hat{n}\right|^2 \right\rangle 2\times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \delta\left(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega\right)$$

Question: What is the real part of the dielectric constant?

Linear Response Functions

Linear Response Functions:

In a linear time invariant (LTI) system, the stimulus phasor $S(\omega)$ is related to the response phasor $R(\omega)$ by a linear response function $\gamma(\omega)$:

$$R(\omega) = \gamma(\omega)\, S(\omega) \qquad \left\{ \gamma(\omega) = \gamma'(\omega) + i\,\gamma''(\omega) \right.$$

The linear system must satisfy the following two properties:

i) It must be causal (system cannot respond before the stimulus is applied)
ii) A real stimulus $S(t)$ must result in a real response $R(t)$ (with no imaginary component)

The second condition gives:

$$\gamma(-\omega) = \gamma^*(\omega) \;\Rightarrow\; \gamma'(-\omega) = \gamma'(\omega) \quad \text{and} \quad \gamma''(-\omega) = -\gamma''(\omega)$$

Most responses of solids are expressed in terms of linear response functions. Examples include:

Conductivity: $\sigma(\omega) \longrightarrow \vec{J}(\vec{r},\omega) = \sigma(\omega)\,\vec{E}(\vec{r},\omega)$

Dielectric Constant: $\varepsilon(\omega) \longrightarrow \vec{D}(\vec{r},\omega) = \varepsilon(\omega)\,\vec{E}(\vec{r},\omega)$

Linear Response Functions and Kramers-Kronig Relations

The two conditions, listed on previous slide, dictate that the real and imaginary parts of any response function cannot be independent – they must be RELATED!

$$R(\omega) = \gamma(\omega)\, S(\omega) \qquad \left\{ \gamma(\omega) = \gamma'(\omega) + i\,\gamma''(\omega) \right.$$

This relationship between the real and the imaginary parts of the response functions is captured by the Kramers-Kronig relations:

$$\gamma''(\omega) = 4\int_0^\infty \frac{d\omega'}{2\pi}\,\left[\gamma'(\omega') - \gamma'(\infty)\right]\frac{\omega}{\omega^2 - \omega'^2} \longrightarrow \quad (1)$$

$$\gamma'(\omega) - \gamma'(\infty) = -4\int_0^\infty \frac{d\omega'}{2\pi}\,\gamma''(\omega')\frac{\omega'}{\omega^2 - \omega'^2} \longrightarrow \quad (2)$$

- If one knows the real part for all frequencies, then one can find the imaginary part using Kramers-Kronig relations

- Conversely, if one knows the imaginary part for all frequencies, then one can find the real part using Kramers-Kronig relations

PROOF OF KRAMERS-KRONIG RELATIONS GIVEN IN APPENDIX

High Frequency Dielectric Constant of Solids: Real Part

We have:

$$\varepsilon''(\omega) = \left(\frac{e}{m}\right)^2 \frac{\pi}{\omega^2} \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \delta\left(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega\right)$$

And from the Kramers-Kronig relations we know:

$$\varepsilon'(\omega) - \varepsilon'(\infty) = -4 \int_0^\infty \frac{d\omega'}{2\pi} \varepsilon''(\omega') \frac{\omega'}{\omega^2 - \omega'^2} \longrightarrow \left\{ \varepsilon'(\infty) = \varepsilon_o \right.$$

$$\Rightarrow \varepsilon'(\omega) - \varepsilon_o = -2\left(\frac{e}{m}\right)^2 \hbar^2 \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \frac{\left(E_c(\vec{k}) - E_v(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_c(\vec{k}) - E_v(\vec{k})\right)^2}$$

$$\Rightarrow \varepsilon'(\omega) = \varepsilon_o - 2\left(\frac{e}{m}\right)^2 \hbar^2 \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \frac{\left(E_c(\vec{k}) - E_v(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_c(\vec{k}) - E_v(\vec{k})\right)^2}$$

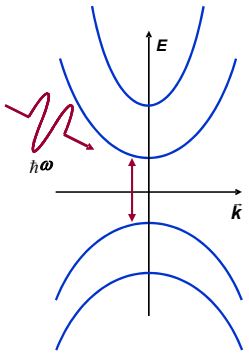
High Frequency Dielectric Constant of Solids

$$\varepsilon'(\omega) = \varepsilon_o - 2\left(\frac{e}{m}\right)^2 \hbar^2 \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \frac{\left(E_c(\vec{k}) - E_v(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_c(\vec{k}) - E_v(\vec{k})\right)^2}$$

$$\varepsilon''(\omega) = \left(\frac{e}{m}\right)^2 \frac{\pi}{\omega^2} \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left[f_v(\vec{k}) - f_c(\vec{k})\right] \delta\left(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega\right)$$

- Note that our expression for dielectric constant takes into account interband transitions involving only a single valence band and a single conduction band

- A more realistic expression would include interband transitions among all bands of the solid

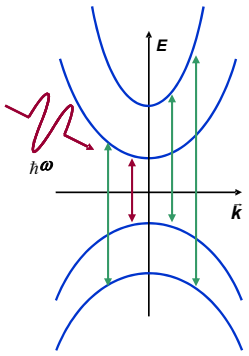


High Frequency Dielectric Constant of Solids

$$\varepsilon''(\omega) = \left(\frac{e}{m}\right)^2 \frac{\pi}{\omega^2} \sum_{r,s} 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left|\vec{P}_{rs} \cdot \hat{n}\right|^2 \left[f_s(\vec{k}) - f_r(\vec{k})\right] \delta\left(E_r(\vec{k}) - E_s(\vec{k}) - \hbar\omega\right)$$

$$\varepsilon'(\omega) = \varepsilon_o - 2\left(\frac{e}{m}\right)^2 \hbar^2 \sum_{r,s} 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left|\vec{P}_{rs} \cdot \hat{n}\right|^2 \left[f_s(\vec{k}) - f_r(\vec{k})\right] \frac{\left(E_r(\vec{k}) - E_s(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_r(\vec{k}) - E_s(\vec{k})\right)^2}$$

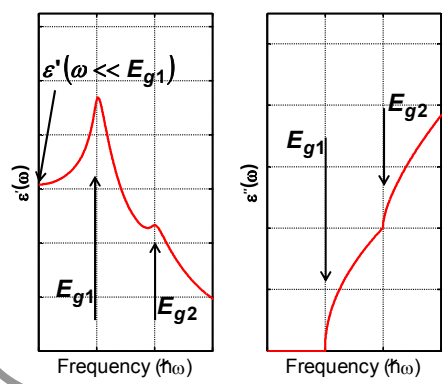
- The above expression includes contributions from interband transitions among all pairs of bands of the solid
- Usually the most important and dominant contribution at frequencies of interest comes from interband transitions between the highest occupied bands (i.e. the valence bands) and the lowest unoccupied band (i.e. the conduction bands)

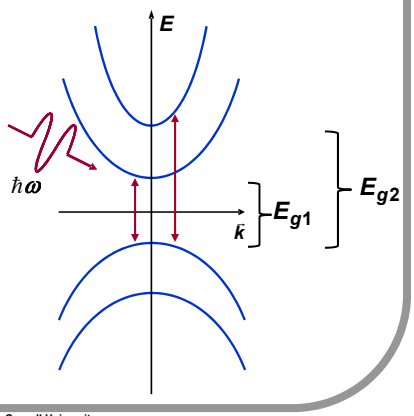


Bandgaps and the High Frequency Dielectric Constant

$$\varepsilon''(\omega) = \left(\frac{e}{m}\right)^2 \frac{\pi}{\omega^2} \sum_{r,s} 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left|\vec{P}_{rs} \cdot \hat{n}\right|^2 \left[f_s(\vec{k}) - f_r(\vec{k})\right] \delta\left(E_r(\vec{k}) - E_s(\vec{k}) - \hbar\omega\right)$$

$$\varepsilon'(\omega) = \varepsilon_o - 2\left(\frac{e}{m}\right)^2 \hbar^2 \sum_{r,s} 2 \times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \left|\vec{P}_{rs} \cdot \hat{n}\right|^2 \left[f_s(\vec{k}) - f_r(\vec{k})\right] \frac{\left(E_r(\vec{k}) - E_s(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_r(\vec{k}) - E_s(\vec{k})\right)^2}$$

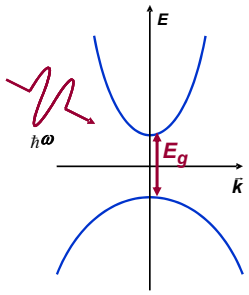




Bandgaps and the High Frequency Dielectric Constant

$$\varepsilon'(\omega) = \varepsilon_o - 2\left(\frac{e}{m}\right)^2 \hbar^2 \left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle \; 2\times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \; \left[f_v(\vec{k}) - f_c(\vec{k})\right] \frac{\left(E_c(\vec{k}) - E_v(\vec{k})\right)^{-1}}{(\hbar\omega)^2 - \left(E_c(\vec{k}) - E_v(\vec{k})\right)^2}$$

Make some very rough estimates:



Suppose : $\hbar\omega << E_g$

$$E_c(\vec{k}) - E_v(\vec{k}) \approx E_g$$

$$f_v(\vec{k}) \approx 1 \qquad f_c(\vec{k}) \approx 0$$

$$\Rightarrow \varepsilon'(\omega) \approx \varepsilon_o + 2\left(\frac{e}{m}\right)^2 \hbar^2 \frac{\left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle}{E_g^3} \; 2\times \int_{FBZ} \frac{d^3\vec{k}}{(2\pi)^3} \; f_v(\vec{k})$$

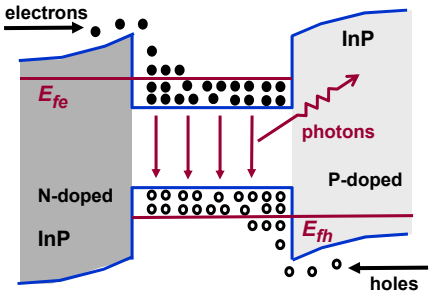
$$\approx \varepsilon_o + \frac{(e\hbar)^2}{2}\left(\frac{1}{m_e} + \frac{1}{m_h}\right)\frac{n_v}{E_g^2}$$

Remember the relation from your homework 7: $\left(\frac{1}{m_e} + \frac{1}{m_h}\right) = \frac{4}{m^2}\frac{\left\langle \left|\vec{P}_{cv} \cdot \hat{n}\right|^2 \right\rangle}{E_g}$

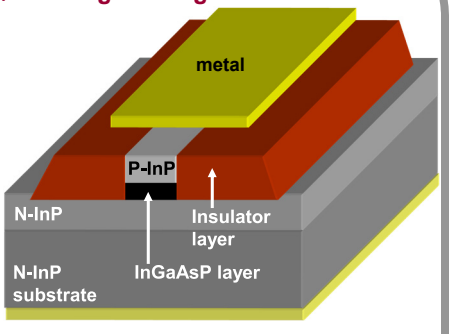
⇒ Materials with larger bandgaps will have smaller real parts of dielectric constants (and, therefore, smaller real parts of refractive indices)

Semiconductor Heterostructure Lasers

A Heterostructure Laser (Band Diagram)



A Ridge Waveguide Laser Structure



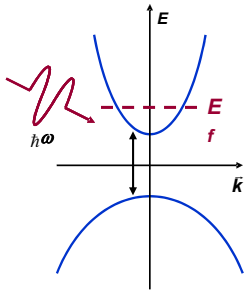
In semiconductor heterostructure laser, the wider bandgap material has smaller refractive index than the narrower bandgap material

The combination of narrow and wide bandgap materials act like a dielectric optical waveguide that confines and guides the photons

The heterostructure not only confines the carriers but it also confines the photons!!

Dielectric Constant: Case of Non-Zero Conductivity

We have obtained an expression for the dielectric constant that incorporated interband optical processes and phonons



What if the material also contained large densities of electrons or holes or both (i.e. what if the material was doped and conductive)?

Go back to Maxwell equations:

$$\nabla \times \vec{E}(\vec{r},\omega) = i\omega \mu_o \vec{H}(\vec{r},\omega)$$ Faraday's Law

$$\nabla \times \vec{H}(\vec{r},\omega) = \vec{J}(\omega) - i\omega \varepsilon(\omega) \vec{E}(\vec{r},\omega)$$ Ampere's Law

New term (current density due to electrons or holes or both)

$$\vec{J}(\omega) = \sigma(\omega) \vec{E}(\omega)$$

$$\Rightarrow \nabla \times \vec{H}(\vec{r},\omega) = \sigma(\omega) \vec{E}(\omega) - i\omega \varepsilon(\omega) \vec{E}(\vec{r},\omega)$$

$$\Rightarrow \nabla \times \vec{H}(\vec{r},\omega) = -i\omega \varepsilon_{\text{eff}}(\omega) \vec{E}(\vec{r},\omega)$$

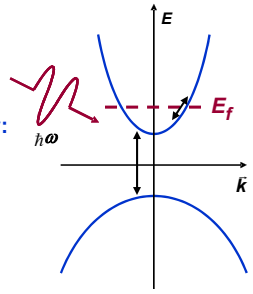
Where:

$$\varepsilon_{\text{eff}}(\omega) = \varepsilon(\omega) + i\frac{\sigma(\omega)}{\omega}$$ → The second term is the intraband or the free-carrier contribution

Dielectric Constant: Non-Zero Conductivity

We have:

$$\varepsilon_{\text{eff}}(\omega) = \varepsilon(\omega) + i\frac{\sigma(\omega)}{\omega}$$



Use the Drude model for the frequency dependent conductivity:

The equation for the electron drift velocity is (assuming parabolic/isotropic bands in 3D):

$$m_e \frac{d\vec{v}}{dt} - \frac{m_e \vec{v}}{\tau} = -e\vec{E}$$

In phasor notation (assuming a sinusoidal electric field):

$$\vec{v}(\omega) = -\frac{e\tau/m_e}{1 - i\omega\tau}\vec{E}(\omega)$$

The current density is:

$$\vec{J}(\omega) = n\, e\, \vec{v}(\omega) = \sigma(\omega)\vec{E}(\omega)$$

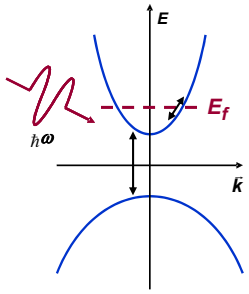
where:

$$\sigma(\omega) = \frac{n\, e^2\tau/m_e}{1 - i\omega\tau}$$

Dielectric Constant: Non-Zero Conductivity

Assuming non-zero densities for both electrons and holes the total conductivity becomes:

$$\sigma(\omega) = \frac{n\, e^2\tau/m_e}{1 - i\omega\tau} + \frac{p\, e^2\tau/m_h}{1 - i\omega\tau}$$



We have:

$$\varepsilon_{eff}(\omega) = \varepsilon(\omega) + i\frac{\sigma(\omega)}{\omega}$$

Therefore:

$$\varepsilon_{eff}(\omega) = \varepsilon(\omega) + i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} + i\frac{pe^2\tau/m_h}{\omega(1 - i\omega\tau)}$$

Interband optical processes and phonons

Intraband optical processes

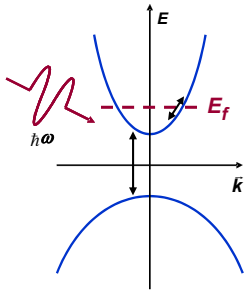
The Plasma Frequency

Suppose we have a metal or a n-doped semiconductor for which:

$$\omega\tau >> 1$$

$$\omega >> \omega_{TO}, \omega_{LO}$$

$$\hbar\omega << E_g$$



Then we have:

$$\varepsilon_{eff}(\omega) = \varepsilon(\omega) + i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} + i\frac{pe^2\tau/m_h}{\omega(1 - i\omega\tau)} \quad (\text{last term} \to 0)$$

$$= \varepsilon(\omega) + i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} \approx \varepsilon(\infty) - \frac{ne^2/m_e}{\omega^2}$$

$$= \varepsilon(\infty)\left(1 - \frac{\omega_p^2}{\omega^2}\right)$$

Where the plasma frequency is defined as:

$$\omega_p = \sqrt{\frac{ne^2}{\varepsilon(\infty)\, m_e}}$$

For metals:

$\omega_p/2\pi \sim 4 \times 10^{15}$ Hz (UV-blue light frequency)

For semiconductors:

$\omega_p/2\pi \sim 10^{11} - 10^{13}$ Hz (Terahertz frequency)

Putting Everything Together

$$\varepsilon_{eff}(\omega) = \varepsilon(\omega) + i\frac{\sigma(\omega)}{\omega} = \varepsilon_o\left(1 + \chi_e(\omega) + \chi_{ph}(\omega)\right) + i\frac{\sigma(\omega)}{\omega}$$

$$= \varepsilon_o + \varepsilon_o\chi_e(\omega) + \varepsilon_o\chi_{ph}(\omega) + i\frac{\sigma(\omega)}{\omega}$$

Electronic part (Interband) | Phononic part | Conductivity part (electronic intraband part or the free carrier part)

Electronic Parts:

$$\varepsilon_o\chi_e(\omega) = -2\left(\frac{e}{m}\right)^2\hbar^2\sum_{r,s}\ 2\times\int_{FBZ}\frac{d^3\vec{k}}{(2\pi)^3}\left|\vec{P}_{rs}\cdot\hat{n}\right|^2\left[f_s(\vec{k}) - f_r(\vec{k})\right]\frac{\left(E_r(\vec{k}) - E_s(\vec{k})\right)^{-1}}{(\hbar\omega + i\eta)^2 - \left(E_r(\vec{k}) - E_s(\vec{k})\right)^2}$$

$$i\frac{\sigma(\omega)}{\omega} = i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} + i\frac{pe^2\tau/m_h}{\omega(1 - i\omega\tau)}$$

First line is the interband part and the second line is the inraband or the free-carrier part

Second line is non-zero only for conducting materials and has no zero frequency limit

Phononic Part:

$$\varepsilon_o\chi_{ph}(\omega) = -\frac{nf^2/M_r}{\omega^2 - \omega_{TO}^2}$$

Polaritons

Polaritons consist of electromagnetic waves coupled with some material wave or material excitation

It is the name given to the phenomena where electromagnetic energy becomes strongly coupled with material degrees of freedom

Some common examples of polaritons are:

1) Phonon-Polaritons

Electromagnetic waves become strongly coupled with the optical phonons of a polar medium

2) Plasmon-Polaritons

Electromagnetic waves become strongly coupled with the plasma waves of a conducting medium

3) Exciton-Polaritons

Electromagnetic waves become strongly coupled with excitons (bound electron-hole pairs)

Transverse and Longitudinal Polaritons

For any medium: $\vec{D} = \varepsilon_o \vec{E} + \vec{P} = \varepsilon(\omega)\vec{E}$ $\rho_p = -\nabla.\vec{P}$

$$\nabla.\vec{E} = \frac{\rho_u + \rho_p}{\varepsilon_o} \qquad \nabla.\vec{D} = \frac{\rho_u}{\varepsilon_o}$$

Longitudinal Polaritons:

In longitudinal polaritons, the E-field has a non-zero divergence but the D-field has a zero divergence:

$$\nabla.\vec{E} = \frac{\rho_u + \rho_p}{\varepsilon_o} = \frac{\rho_p}{\varepsilon_o} \neq 0 \qquad \nabla.\vec{D} = \frac{\rho_u}{\varepsilon_o} = 0 \quad \Rightarrow \quad \rho_p = -\nabla.\vec{P} \neq 0$$

If the E-field has a wave-like form: $\vec{E} = \hat{n}E_o e^{i\vec{q}.\vec{r} - i\omega t}$

Then: $\nabla.\vec{E} \neq 0 \quad \Rightarrow \quad \vec{q}.\hat{n} \neq 0$

$\Rightarrow$ E-field has a non-zero component in the direction of wave propagation

Transverse Polaritons:

In transverse polaritons, the E-field and the D-field both have a zero divergence:

$$\nabla.\vec{E} = 0 \qquad \nabla.\vec{D} = 0 \quad \Rightarrow \quad \rho_p = 0$$

If the E-field has a wave-like form: $\vec{E} = \hat{n}E_o e^{i\vec{q}.\vec{r} - i\omega t}$

Then: $\nabla.\vec{E} = 0 \quad \Rightarrow \quad \vec{q}.\hat{n} = 0$

$\Rightarrow$ E-field has no component in the direction of wave propagation

Longitudinal Polaritons

Suppose the E-field has a wave-like form:

$$\vec{E} = \hat{n}E_o e^{i\vec{q}.\vec{r} - i\omega t}$$

The D-field is given as:

$$\vec{D} = \varepsilon_o \vec{E} + \vec{P} = \varepsilon_{eff}(\omega)\vec{E}$$

For longitudinal polaritons we must have:

$$\nabla.\vec{E} \neq 0$$

$$\nabla.\vec{D} = \varepsilon_{eff}(\omega)\nabla.\vec{E} = 0$$

The only way that both these equations can hold is if the frequency of the longitudinal polaritons is such that:

$$\varepsilon_{eff}(\omega) = 0$$

The above equation gives the frequency of the longitudinal polaritons

Longitudinal Polaritons

Longitudinal Phonon-Polaritons: Consider a non-conducting polar medium (polar semiconductor or a polar insulator) whose dielectric constant at frequencies much smaller than the material bandgap energies is approximately,

$$\varepsilon_{eff}(\omega) = \varepsilon(\infty) - \frac{nf^2/M_r}{\omega^2 - \omega_{TO}^2} = \varepsilon(\infty) - \omega_{TO}^2 \frac{(\varepsilon(0) - \varepsilon(\infty))}{\omega^2 - \omega_{TO}^2} = \varepsilon(\infty)\left[\frac{\omega^2 - \omega_{LO}^2}{\omega^2 - \omega_{TO}^2}\right]$$

The condition, $\varepsilon_{eff}(\omega) = 0$ gives:

$$\omega = \omega_{LO}$$

The longitudinal phonon-polaritons are just the polar longitudinal optical phonons!

Longitudinal Plasmon-Polaritons: Consider a conducting medium (like gold, silver) whose dielectric constant at frequencies much larger than the phonon frequencies but much smaller than the material bandgap energies is approximately,

$$\varepsilon_{eff}(\omega) = \varepsilon(\infty) + i\frac{\sigma(\omega)}{\omega} = \varepsilon(\infty) + i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} \approx \varepsilon(\infty) - \frac{ne^2/m_e}{\omega^2} \qquad \{\omega\tau >> 1$$

The condition, $\varepsilon_{eff}(\omega) = 0$ gives:

$$\omega = \omega_p = \sqrt{\frac{ne^2}{\varepsilon(\infty)m_e}}$$

The longitudinal plasmon-polaritons are just the plasma waves!

Transverse Polaritons

Suppose the E-field has a wave-like form:

$$\vec{E} = \hat{n}E_o e^{i\vec{q}.\vec{r} - i\omega t} \qquad \{\nabla.\vec{E} = 0 \quad \Rightarrow \quad \vec{q}.\hat{n} = 0$$

The D-field is given as:

$$\vec{D} = \varepsilon_o\vec{E} + \vec{P} = \varepsilon_{eff}(\omega)\vec{E}$$

For transverse polaritons we must have:

$$\nabla.\vec{E} = \nabla.\vec{D} = 0$$

The electromagnetic wave equation when $\nabla.\vec{E} = 0$ is:

$$\nabla \times \nabla \times \vec{E} = \omega^2\mu_o\varepsilon_{eff}(\omega)\vec{E}$$
$$\Rightarrow \nabla\underbrace{(\nabla.\vec{E})}_{0} - \nabla^2\vec{E} = \omega^2\mu_o\varepsilon_{eff}(\omega)\vec{E}$$
$$\Rightarrow -\nabla^2\vec{E} = \omega^2\mu_o\varepsilon_{eff}(\omega)\vec{E}$$

The plane wave is a solution of the wave equation if:

$$\omega^2\frac{\varepsilon_{eff}(\omega)}{\varepsilon_o} = q^2c^2$$

The above equation gives the dispersion of the transverse polaritons

Transverse Phonon-Polaritons

Consider a non-conducting polar medium (polar semiconductor or a polar insulator) whose dielectric constant at frequencies much smaller than the material bandgap energies is approximately,

$$\varepsilon_{eff}(\omega) = \varepsilon(\infty) - \frac{nf^2/M_r}{\omega^2 - \omega_{TO}^2} = \varepsilon(\infty) - \omega_{TO}^2 \frac{(\varepsilon(0) - \varepsilon(\infty))}{\omega^2 - \omega_{TO}^2} = \varepsilon(\infty)\left[\frac{\omega^2 - \omega_{LO}^2}{\omega^2 - \omega_{TO}^2}\right]$$

The dispersion relation: $\omega^2 \frac{\varepsilon_{eff}(\omega)}{\varepsilon_o} = q^2c^2$

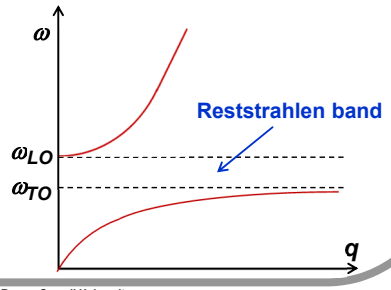
gives the following equation:

$$\omega^4 - \omega^2\left(\omega_{LO}^2 + q^2c^2\,\varepsilon_o/\varepsilon(\infty)\right) + q^2c^2\omega_{TO}^2\,\varepsilon_o/\varepsilon(\infty) = 0$$

The resulting dispersion relation is plotted in the Figure

Note that there is a band of frequencies in which no electromagnetic wave can propagate in the medium
(no propagating wave mode exists)

This band is called the Restsrahlen band



Transverse Plasmon-Polaritons

Consider a conducting medium (like gold, silver) whose dielectric constant at frequencies much larger than the phonon frequencies but much smaller than the material bandgap energies is approximately,

$$\varepsilon_{eff}(\omega) = \varepsilon(\infty) + i\frac{\sigma(\omega)}{\omega} = \varepsilon(\infty) + i\frac{ne^2\tau/m_e}{\omega(1 - i\omega\tau)} \approx \varepsilon(\infty) - \frac{ne^2/m_e}{\omega^2} \qquad \{\omega\tau >> 1$$

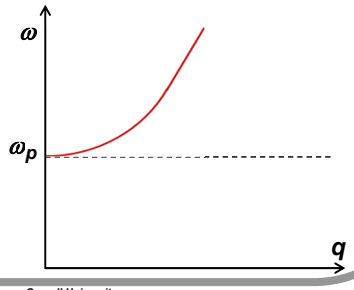
The dispersion relation: $\omega^2 \frac{\varepsilon_{eff}(\omega)}{\varepsilon_o} = q^2c^2$

gives the following equation:

$$\omega^2 = \omega_p^2 + q^2c^2\frac{\varepsilon_o}{\varepsilon(\infty)} \qquad \left\{\omega_p = \sqrt{\frac{ne^2}{\varepsilon(\infty)m_e}}\right.$$

The resulting dispersion relation is plotted in the Figure

Note that no electromagnetic wave can propagate in the medium with a frequency smaller than the plasma frequency



APPENDIX: Kramers-Kronig Relations (Proof)

In a linear time invariant (LTI) system, the stimulus phasor $S(\omega)$ is related to the response phasor $R(\omega)$ by:

$$R(\omega) = \gamma(\omega)\, S(\omega)$$

The linear response function is $\gamma(\omega)$: $\gamma(\omega) = \gamma'(\omega) + i\,\gamma''(\omega)$

Reality:

Real inputs must result in a real response. This condition gives:

$$\gamma(-\omega) = \gamma^*(\omega) \;\Rightarrow\; \gamma'(-\omega) = \gamma'(\omega) \quad \text{and} \quad \gamma''(-\omega) = -\gamma''(\omega)$$

Causality:

Inverse FT gives: $R(t) = \int_{-\infty}^{\infty} dt'\; \gamma(t-t')\, S(t')$ $\qquad \left\{ \gamma(t-t') = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi}\; \gamma(\omega)\, e^{-i\,\omega\,(t-t')} \right.$

Causality implies that the system cannot exhibit response to an input before the input occurs:

$$\gamma(t-t') = 0 \qquad \text{for} \quad t < t'$$

which gives: $R(t) = \int_{-\infty}^{t} dt'\; \gamma(t-t')\, S(t')$

Infinite Frequency Response:

No physical system can respond at infinite frequencies, so:

$$\gamma(\omega \to \infty) = 0$$

Kramers-Kronig Relations (Proof)

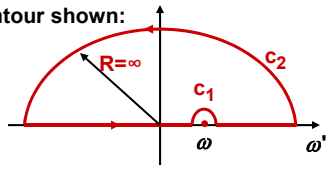
The causality condition is:

$$\gamma(t) = 0 \qquad \text{for} \quad t < 0$$

The function $\gamma(\omega)$, when considered as an analytic function in the complex plane, cannot have any pole in the upper half of the complex plane for the causality condition to hold

Consider the following contour integral over the contour shown:

$$\oint \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'}$$



Since there are no poles in the upper half plane, the closed contour contains no poles, and the contour integral must be zero

$$\oint \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'} = 0$$

$$\Rightarrow \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'} + \int_{c_1} \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'} + \int_{c_2} \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'} = 0 \quad (\text{the } c_2 \text{ integral} \to 0)$$

$$\Rightarrow \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi}\, \frac{\gamma(\omega')}{\omega - \omega'} + \frac{i}{2}\gamma(\omega) = 0$$

Kramers-Kronig Relations (Proof)

$$\Rightarrow \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\gamma(\omega')}{\omega - \omega'} = -\frac{i}{2}\gamma(\omega)$$

Matching the real and imaginary parts on both sides gives:

$$\gamma'(\omega) = -2\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\gamma''(\omega')}{\omega - \omega'} = -4\int_{0}^{\infty} \frac{d\omega'}{2\pi} \gamma''(\omega') \frac{\omega'}{\omega^2 - \omega'^2}$$

$$\gamma''(\omega) = 2\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\gamma'(\omega')}{\omega - \omega'} = 4\int_{0}^{\infty} \frac{d\omega'}{2\pi} \gamma'(\omega') \frac{\omega}{\omega^2 - \omega'^2}$$

Where the following relations have been used to get the second integrals:

$$\gamma'(-\omega) = \gamma'(\omega) \quad \text{and} \quad \gamma''(-\omega) = -\gamma''(\omega)$$

In cases where the real part of $\gamma(\omega)$ may not be zero at infinite frequencies, as it happened in the case of the dielectric constant, we just repeat the entire procedure from the beginning with $\gamma(\omega) - \gamma'(\infty)$ instead of $\gamma(\omega)$ to get:

$$\gamma''(\omega) = 4\int_{0}^{\infty} \frac{d\omega'}{2\pi} \left[\gamma'(\omega') - \gamma'(\infty)\right] \frac{\omega}{\omega^2 - \omega'^2}$$

$$\gamma'(\omega) - \gamma'(\infty) = -4\int_{0}^{\infty} \frac{d\omega'}{2\pi} \gamma''(\omega') \frac{\omega'}{\omega^2 - \omega'^2}$$